

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF

SERIAL NO.: 10/501,100

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FOR: NI BASED ALLOY WITH EXCELLENT CORROSION RESISTANCE TO
SUPERCRITICAL WATER ENVIRONMENTS CONTAINING INORGANIC ACIDS

EXAMINER: Roy King

GROUP ART UNIT: 1793

DECLARATION UNDER 37 C.F.R. § 1.132

ASSISTANT COMMISSIONER FOR PATENTS

ALEXANDRIA, VIRGINIA 22313-1450

SIR:

I, Katsuo Sugahara, a Japanese citizen residing at 2-1-3 Motomachi, Urawa-ku,
Saitama-shi, Saitama, 330-0073, declare and state that:

I obtained my doctor's degree in metallurgical engineering from Tokyo
Institute of Technology in March, 1998.

I entered Mitsubishi Materials Corporation in April 1 1990.

I am one of the Applicants of the above-identified application and I am well
familiar with the present case.

I have a good knowledge of the English language and have read and
understood the application papers and the prosecution history of this and the antecedent
applications as well as the Examiner's references cited therein.

PURPOSE, METHOD AND RESULTS

I. Purpose of the experiments:

The purpose of the Experiments described in this declaration is to show that it is indispensable that all of Cr, Fe and Si are within the range of claim 2 in order to achieve the present invention, and that, if any of Cr, Fe and Si is not within the range of claim 2, the corrosion resistance deteriorates in supercritical water environments containing inorganic acids. Furthermore, another purpose thereof is to show the criticality of the amount in the range of Cr, Fe, and Si.

II. Method of the Experiments:

Using a raw material with a low C content in each case, the raw material was melted and cast in a normal high frequency induction furnace to prepare an ingot of 12mm in thickness. The ingot was then subjected to homogenizing heat treatment for 10 hours at 1230°C. Subsequently, with the temperature held within a range from 1000°C to 1230°C, hot rolling was used to reduce the thickness by 1 mm per repetition, and this process was repeated until a final thickness of 5 mm was achieved. The sample was then subjected to solution treatment by holding the sample at 1200°C for 30 minutes followed by water quenching. The surface of the sample was then buffed, yielding a Ni based alloy sheet of "Sample No. 1 to 20".

Each of the Ni based alloy sheets of "Sample No. 1 to 20" was cut to prepare solution test specimens of dimensions 10 mm x 50 mm. In addition, in order to evaluate the effect of the phase stability on the corrosion resistance relative to a supercritical water environment containing inorganic acids, each of the Ni base alloy sheets of "Sample No. 1 to 20" was subjected to aging treatment by holding the sheet at 550°C for 1000 hours, and the sheet was then cut to prepare aged test specimens of dimensions 10 mm x 50 mm.

Next, a flow type corrosion test apparatus was prepared using a Hastelloy C-276 pipe as an autoclave. A test solution is pumped into one end of the Hastelloy C-276 pipe of this flow type corrosion test apparatus using a high pressure pump, and is discharged from the other end of the pipe, while the test solution inside the Hastelloy C-276 pipe is maintained at a predetermined flow rate. The test solution is heated by a heater provided on the Hastelloy C-276 pipe, and the test solution is able to be maintained at a predetermined temperature. In addition, the test solution discharged from the other end of the Hastelloy C-276 pipe of the flow type corrosion test apparatus passes through a pressure reducing valve and is recovered in a reservoir tank.

Using the flow type corrosion test apparatus described above, corrosion tests were conducted using the inorganic acid containing supercritical water simulated solutions described below.

(Aa) A test solution was prepared by mixing 0.2 mol/kg of sulfuric acid and 0.2 mol/kg of phosphoric acid into supercritical water with a fluid temperature of 550°C, a pressure of 40 MPa and a dissolved oxygen level of 8 ppm. This solution is an estimation of the supercritical water solution generated when VX gas is decomposed and oxidized in supercritical water (and is hereafter referred to as a simulated VX gas decomposition supercritical water solution). This simulated VX gas decomposition supercritical water solution was fed into the Hastelloy C-276 pipe of the aforementioned flow type corrosion test apparatus, and the flow rate of the simulated VX gas decomposition supercritical water solution inside the Hastelloy C-276 pipe was adjusted to 6 g/min, thus forming a supercritical water environment containing inorganic acids. Solution test specimens of the Ni based alloy sheets of "Sample No. 1 to 20" were then each held in this supercritical water environment for a period of 100 hours. The reduction in weight of the solution test specimen over the course of the test was divided by the surface area of the specimen to determine the weight loss per unit area for each test specimen.

In addition, in order to evaluate the effect of the phase stability on the corrosion resistance relative to a supercritical water environment containing inorganic acids, aged test specimens of the Ni based alloy sheets of "Sample No. 1 to 20" were each held in the above supercritical water environment containing inorganic acids for a period of 100 hours. The reduction in weight of the test specimen over the course of the test was divided by the surface area of the aged test specimen to determine the weight loss per unit area for each test specimen.

(Ab) A test solution was prepared by mixing 0.4 mol/kg of phosphoric acid and 0.1 mol/kg of hydrofluoric acid into supercritical water with a fluid temperature of 550°C, a pressure of 40 MPa and a dissolved oxygen level of 8 ppm. This solution is an estimation of the supercritical water solution generated when GB (sarin) gas is decomposed and oxidized in supercritical water (and is hereafter referred to as a simulated GB gas decomposition supercritical water solution). The simulated GB gas decomposition supercritical water solution was fed into the Hastelloy C-276 pipe of the aforementioned flow type corrosion test apparatus, and the flow rate of the simulated GB gas decomposition supercritical water solution inside the Hastelloy C-276 pipe was

adjusted to 6 g/min, thus forming a supercritical water environment containing inorganic acids. Solution test specimens of the Ni based alloy sheets of "Sample No. 1 to 20" were then each held in this supercritical water environment for a period of 100 hours. The reduction in weight of the solution test specimen over the course of the test was divided by the surface area of the specimens to determine the weight loss per unit area for each test specimen.

In addition, in order to evaluate the effect of the phase stability on the corrosion resistance relative to a supercritical water environment containing inorganic acids, aged test specimens of the Ni based alloy sheets of "Sample No. 1 to 20" were each held in the above supercritical water environment containing inorganic acids for a period of 100 hours. The reduction in weight of the test specimen over the course of the test was divided by the surface area of the aged test specimen to determine the weight loss per unit area for each test specimen.

III. Results:

The components (% by weight) of Sample No. 1 to 20 and the results are shown below in Table 1.

Please pay attention especially to Cr, Fe and Si in the components of Sample No. 1 to 20.

[Table 1]

	Sample No.	Composition (% by weight)							Corrosion tests using simulated V-X gas decomposition supercritical water solution			Corrosion tests using simulated GB gas decomposition supercritical water solution		
		Cr	Mo	Mg	N	Mn	Fe	Si	C ⁺	Ni and unavoidable impurities	weight reduction in solution test specimen (mg/cm ²)	weight reduction in aged test specimen (mg/cm ²)	weight reduction in solution test specimen (mg/cm ²)	weight reduction in aged test specimen (mg/cm ²)
Previously submitted examples	1	44.5	0.80	0.07	0.018	0.15	-	0.33*	0.02	remainder	13	35	16	33
	2	39.8*	0.78	0.08	0.008	0.18	-	-	0.02	remainder	22	25	29	32
	3	44.8	0.61	0.07	0.009	0.11	3.7*	-	0.02	remainder	19	23	21	24
	4	39.9*	0.51	0.08	0.015	0.22	3.3*	0.32*	0.02	remainder	24	35	32	41
	5	42.8*	0.26	0.005	0.009	0.08	-	0.04	0.02	remainder	13	14	14	15
Newly added examples	6	42.6*	0.39	0.012	0.012	0.07	0.35	-	0.02	remainder	14	15	15	15
	7	42.7*	0.65	0.016	0.018	0.07	0.31	0.03	0.02	remainder	13	14	13	14
	8	43.2	0.37	0.021	0.011	0.08	7.63*	-	0.02	remainder	12	13	13	15
	9	43.5	0.41	0.025	0.015	0.07	7.13*	0.03	0.02	remainder	14	15	15	16
	10	43.6	0.46	0.027	0.010	0.07	-	0.12*	0.02	remainder	9	15	10	17
	11	43.3	0.37	0.021	0.017	0.09	0.27	0.13*	0.01	remainder	8	14	10	16
	12	42.7*	0.35	0.019	0.016	0.09	7.63*	-	0.02	remainder	16	17	18	16
	13	42.6*	0.41	0.018	0.018	0.08	-	0.17*	0.02	remainder	14	18	15	19
	14	43.4	0.34	0.023	0.022	0.07	1.04*	0.12*	0.02	remainder	14	18	14	19
	15	42.9*	0.31	0.015	0.025	0.07	1.02*	0.13*	0.02	remainder	17	19	18	20
	16	43.7	0.43	0.017	0.014	0.08	0.95	-	0.02	remainder	7	8	8	8
	17	43.6	0.39	0.014	0.019	0.09	0.96	0.08	0.02	remainder	8	9	8	9
	18	43.5	0.46	0.026	0.026	0.08	-	0.08	0.02	remainder	7	8	8	8
	19	43.8	0.44	0.021	0.024	0.07	0.18	0.09	0.02	remainder	6	8	8	8
	20	43.6	0.48	0.019	0.020	0.09	0.94	0.02	0.02	remainder	7	8	8	9

(1) The results of Sample No. 5 to 7 :

In Samples No. 5 to 7, the amount of Cr was 42.8% by weight, 42.6% by weight, and 42.7% by weight, respectively, which are not within the range of claim 2 (more than 43% to 50% or less), but within the range of Kazuo Yamanaka et al. (38 to 45%), whereas the amount of other components were within the range of claim 2.

The results indicate that, if the content of Cr is beyond the range of claim 2, that is, if the quantity of Cr is 43% or less, the corrosion resistant effect cannot be achieved.

(2) The results of Sample No. 8 and 9:

In Samples No. 8 and 9, the content of Fe was 1.05% by weight and 1.13% by weight, respectively, which are not within the range of claim 2 (0.05 to 1%), but within the range of Kazuo Yamanaka et al. (greater than 0 to 21.5% according to the Examiner's opinion), whereas the contents of other components are within the range of claim 2.

The results indicate that the content of Fe beyond the range of claim 2, that is, quantities exceeding 1%, result in an undesirable deterioration in the corrosion resistance relative to supercritical water environments containing inorganic acids.

(3) The results of Sample No. 10 and 11:

In Samples No. 10 and 11, the content of Si was 0.12% by weight and 0.13% by weight, respectively, which are not within the range of claim 2 (0.01 to 0.1%), but within the range of Kazuo Yamanaka et al. (from some extent to 1%, see Examples), whereas the contents of other components were within the range of claim 2.

The results indicate that the content of Si beyond the range of claim 2, that is, quantities exceeding 0.1%, results in an undesirable deterioration in the corrosion resistance relative to supercritical water environments containing inorganic acids.

(4) The results of Sample No. 12 to 15:

In Samples No. 12 to 15, the amount of two or three of Cr, Fe and Si was not within the range of the present invention, but within the range of Kazuo Yamanaka et al., whereas the contents of other components are within the range of the present invention.

In the case that two or all of Cr, Fe and Si were not within the range of claim 2, the corrosion resistance in supercritical water environments containing inorganic acids became much worse than in the case of Samples No. 5 to 11, in which the amount of

one of Cr, Fe, and Si is outside the range of claim 2. Therefore, it is necessary that all of Cr, Fe and Si are within the range of claim 2 in order to achieve the present invention.

IV. Conclusion:

For these reasons, it can be fairly concluded that the present invention has a distinguished effect over Kazuo Yamanaka et al. by containing Cr, Fe and Si within the range of claim 2, not the range of Kazuo Yamanaka et al. Also, it is confirmed that, if even one of the components is beyond the range of claim 2, the present invention cannot be achieved.

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed true; and further that these statements were made with knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Katsuo Sugahara

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